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FINAL REPORT

Modeling, Simulation and Engineering Scale-up Procedures for Design of CVD Reactors

Contract Title: Modeling, Simulation and Engineering Scale-up
Procedures for the Design of CVD Reactors

Performing Organization: Laboratory for Ceramic and Reaction Engineering
Department of Chemical Engineering
State University of New York at Buffalo

Principal Investigator: Vladimir Hlavacek

Contract Number: DU01U0770

R. & T Project Number: N00014-89-J3203

ONR Scientific Officer: Dr. Robert C. Pohanka

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Description of the Scientific Research Goals

The research program, initiated in 1989 by the Laboratory for Ceramic and Reaction Engineering (LCRE), was aimed towards the development of generic processes for growing thick films deposited by CVD techniques. The application of thick-film technology includes the manufacturing of optical windows and ceramic plates, fabrication of optical domes and refractory crucibles, production of refractory metal tubes, ceramic fibers, etc. The objective of this research was to understand the complex phenomena which could occur in a CVD system, including problems of reactive fluid flow, instability of the deposition process at the interphase fluid-solid, stability of the nucleation process on the solid surface, and generation and development of stresses due to thermal gradients and growth-induced mechanisms. New ways of increasing the rate of deposition were also contemplated. Scale-up of CVD reactors for the applications previously mentioned, has not received much attention in the literature. LCRE has since been working on such Chemical Engineering procedures as the gathering and measuring of necessary data, and a priori simulation of laboratory bench-scale units and scale-up to pilot-plant size.

Summary of Progress

Both experimental and theoretical research were performed under this program. The theoretical research consisted mainly in the development of software for simulation and design of CVD reactors for the coating of fibrous substrates. The salient features of the software developed during this period are:

- Two-dimensional and fully three-dimensional simulations.
- Additional transport mechanisms have been incorporated (Soret effect, radiation).
- Different kinetic mechanism have been analyzed.
- Bulk and fiber-scale phenomena have been analyzed independently.

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- Steady-state and dynamic results have been obtained.
- Development of software for the analysis of different CVD applications (i.e., ceramic tubes and plates).
- Implementation of a code for the prediction of phase diagrams in different systems of interest to the manufacture of ceramic tubes and plates by CVD.
- Development of the routines for transport and physico-chemical parameter estimation. These routines are part of a modular package for the analysis of CVD systems.
- Analysis of thermal and growth-induced stresses in the deposition of thick ceramic coatings (titanium diboride mainly).
- Implementation of additional graphics and animation techniques for a better understanding of the underlying physics in CVD processes (i.e., visualization of the results of numerical simulations).

The experimental investigation has been concentrated in the production of thick films and its application in the manufacture of ceramic tubes. A summary of the experimental research is as follows:

- Different reaction systems have been analyzed (i.e., silicon carbide, boron, and titanium diboride coatings on graphite tubes).
- Horizontal arrangements have been investigated achieving results for silicon carbide tubes (3-10 mm thick smooth coatings).
- A technological process has been developed to produce SiC tubes from a CVD process.
- Design and construction of different CVD bench-scale reactors.
- Development of techniques of very fast CVD.

List of Publications/Reports/Presentations

1. Papers Published in Refereed Journals

Scholtz, J.H., J.E. Gatica, H.J. Viljoen and V. Hlavacek, "Coating of Fibrous Substrates by CVD. Analysis of the Fiber Evolution," *J. Crystal Growth* 108, pp. 190-202 (1991).

Scholtz, J.H., V. Lakhota, J.E. Gatica, V. Revankar and V. Hlavacek, "High Performance Ceramic Fibers by CVD Technology," *Materials and Manufacturing Processes*, special issue on CVD Processing 6 (3), 397 (1991).

Scholtz, J.H., J.E. Gatica, H.J. and V. Hlavacek, "Analysis of Transport Phenomena in the Coating of Fibers by CVD," to appear, *Les Editions de Physique*, Proceedings of the 8th European CVD Meeting, Glasgow, UK (1991).

Scholtz, J.H., J.E. Gatica, H.J. Viljoen and V. Hlavacek, "The Coating of Fibrous Substrates by CVD. Modeling and Simulation," Feature article, to appear *Amer. Cer. Soc. J.*, (1993).

Scholtz, J.H., Gatica, J.E., Viljoen, H.J., Revankar, V., and V. Hlavacek, "CVD Reactors for the Synthesis of Inorganic Fibers Modeling and Experimental Evaluation," *Chem. Eng. Sci.*, 45, 2543 (1990).

Hlavacek, V., Scholtz, J. H.: "CVD Reactors for the Coating of Fibrous Substrates. Modeling and Numerical Simulation," *J. Electrochem. Soc.* 137, 3459 (1990).

Revankar, V., Hlavacek V.: "Multifilament Silicon Carbide Fibers by Chemical Vapor Deposition (CVD)," *High Temperature Materials and Processes* 10, 55 (1991).

Revankar, V., Hlavacek V.: "Intermetallic Matrix Reinforcement Ceramic Fibers by Chemical Vapor Deposition," *High Temperature Materials and Processes*, 10, 43 (1991).

Viljoen, H. J., Hlavacek, V.: "Temperature Oscillations and Vibrations of Alternating Current Resistively Heated Thin Filaments," *AIChE Jour.*, 38, 284 (1992).

Viljoen, H. J., Hlavacek, V.: "Temperature Oscillations of Alternating-Current-Heated Thin Filaments in Ceramic Fiber Production," *J. Am. Cer. Soc.*, 75, 1997 (1992).

Orlicki, D., Hlavacek, V., Viljoen, H. J.: "Modeling of a Si:H Deposition in a dc Glow Discharge Reactor," *J. Mat. Research* 7, 8, 2160 (1992).

Zhao, G. Y., Zhu, Ch.W., Revankar, V., Hlavacek, V.: "Silicon Carbide Tubes by Chemical Vapor Deposition Technology," *Jour. of Mat. Synth. and Processing*, 1, 159 (1993).

Scholtz, J. H., Gatica, J. E., Viljoen, H. J., Hlavacek, V.: Analysis of Transport Phenomena in the Coating of Fibers by CVD," *J. Phys. IV*, 1, C2/135-C2/142 (1991).

2. Non-Refereed Publications and Published Technical Reports

Revankar, V., Scholtz, J., Hlavacek, V.: "Synthesis of High Performance Ceramic Fibers by Chemical Vapor Deposition: A Modeling Study," *Cer. Eng. Sci. Proces.* 2, [7-8], 919 (1990).

Revankar, V., Lakhota, V., Hlavacek, V.: "Material Characterization of Chemical Vapor Deposited TiB₂ Fibers," *Cer. Eng. Sci. Proc.*, 12 [7-8], 1086 (1991).

3. Presentations

a. Invited

Scholtz, J. H., Gatica, J. E., Viljoen, H. J., Revankar, V., Hlavacek, V.: "CVD Reactors for the Synthesis of Inorganic Fibers, ISCRE 11 (Toronto, July 8-1, 1990).

Scholtz, J. H., Gatica, J. E., Viljoen, H. J., Hlavacek, V.: "Analysis of Transport Phenomena in the Coating of Fibers by CVD," 8th European CVD Symposium (Glasgow, Scotland, Sept. 9-13, 1991).

b. Contributed

Scholtz, J. H., J. E. Gatica, H. J. Viljoen and V. Hlavacek: "CVD Technologies for the Coating of Fibrous Substrates. Modeling and Numerical Simulation," Annual AIChE Meeting, Chicago, IL (1990).

Scholtz, J. H. J. E. Gatica, H. J. Viljoen and V. Hlavacek, "Thermal Stresses in Ceramic Fibers Manufactured by CVD Technologies," 15th Annual Conference on Composites and Advanced Ceramics, Cocoa Beach, FL (1991).

Schmidt, W. U., V. Revankar and V. Hlavacek, "A Thermodynamic and Experimental Analysis of CVD Deposited Chromium Diboride Fibers Using Chromyl Chloride Precursor," 15th Annual Conference on Composites. Materials and Structures - restricted sessions, Cocoa Beach, FL (1991).

Viljoen, H. J., Wright, C. J., Gatica, J. E., Hlavacek, V.: "Stability of Compressible Fluids in Horizontal CVD Reactors," Annual AIChE Meeting (San Francisco, 1989).

Scholtz, J. H., Gatica, J. E., Viljoen, H. J., Hlavacek, V.: "CVD Reactors for the Coating of Fibrous Substrates, Evolution of the Fiber," Amer. Cer. Soc. Mtg. (22-26 April, 1990, Dallas).

Lakhota, V., Revankar, V., Hlavacek, V.: "Material Characterization of Chemical Vapor Deposited TiB₂ Fibers," Amer. Cer. Soc. Mtg. (April 22-26, 1990, Dallas).

Hlavacek, V., Zhu, Ch.W., Zhao, G. Y.: "Silicon Carbide Tubes Via CVD," Annual AIChE Meeting, Nov. 1-6, 1992, Miami Beach, FL

Thiart, J. J., Viljoen, H. J., Wright, C., Hlavacek, V.: "Analysis of the Morphological Stability of CVD Fibers," Annual AIChE Meeting, Nov. 1-6, 1992, Miami Beach, FL

List of Honors/Awards

<u>Name of Person Receiving Award</u>	<u>Recipient's Institution</u>	<u>Name, Sponsor and Purpose of Award</u>
Vladimir Hlavacek	State University of New York at Buffalo	Clifford C. Furnas Eminent Professor of Chemical Engineering
Vladimir Hlavacek	State University of New York at Buffalo	R. G. Wilhelm Award The American Institute of Chemical Engineers for distinguished and continuing contribution to Chemical Reaction Engineering

Project participants and their status

<u>Name</u>	<u>Status</u>
Jan A. Puszynski	Research Associate Professor
Vithal Revankar	Postdoctoral Associate
Guo-Ying Zhao	Postdoctoral Associate
Ching-Wen Zhu	Postdoctoral Associate
Dariusz Orlicki	Postdoctoral Associate
Hendrik J. Viljoen	Postdoctoral Associate
Jorge E. Gatica	Postdoctoral Associate
Jan H. Scholtz	Graduate Student
Wolfgang U. Schmidt	Graduate Student
Hilmar Rode	Graduate Student
Johannes J. Thiart	Graduate Student

Degrees granted

<u>Name</u>	<u>Degree</u>
Jan H. Scholtz	Doctor of Philosophy
Wolfgang U. Schmidt	Masters of Science